

Amendment  
Application No. 10/602,906  
Attorney Docket No. 052455

### **REMARKS**

Claims 1-11 are pending in this application, of which claims 1, 5 and 10 have been amended, and claims 12-16 have been added.

Claim 5 has been amended, whose basis is found at Examples 1-5 of the present invention. The content of the water-soluble organic high molecular weight compound based on the total solution in Examples 1-5 is calculated as follows: Example 1: 1.00 wt%; Example 2: 1.06 wt%; and Example 3-5: 1.54 wt%. Since original claim 5 describes that the water-soluble organic high molecular weight compound is present in a ratio of 0.03 to 10wt% based on the total solution, and since Examples support the content of 1.00 wt% or higher, the amendment of claim 5 is supported. *In re Wertheim*, 541 F.2d 257, 191 USPQ 90 (CCPA 1976). MPEP2163.05.

Newly added claim 12 is supported at page 23, lines 6-11; newly added claim 13 is supported at page 27, lines 13-14; newly added claim 14 is supported at page 23, lines 2-4; newly added claim 15 is supported at page 27, line 18 to page 28, line 3; and newly added claim 16 is supported at page 30, lines 7-10.

- (1) Claims 1-11 were rejected under 35 U.S.C. §112, second paragraph, as being indefinite.

Amendment  
Application No. 10/602,906  
Attorney Docket No. 052455

In accordance with the Examiner's suggestion, claim 1 has been amended to change "water-soluble organic high molecular compound" to "water soluble organic high molecular weight compound," as supported on page 30, lines 11-21.

(2) Claims 1-7, 10 and 11 were rejected under 35 U.S.C. §103(a) as being unpatentable over Olson (U.S. Patent No. 2,617,742) and in view of Furusaki et al. (Furusaki et al., "Preparation of ITO Thin Film by Sol-Gel Method," Journal of the Ceramic Society of Japan, 102(2), pp 200-205 (1994)).

Applicants herewith file a partial translation of Furusaki et al.

(i) As to claim 1, Olson discloses transparent electroconductive films prepared by applying a solution to a lime soda glass substrate, the solution containing water-soluble indium compound (indium chloride), halogen-containing water-soluble organotin compound (ethyl tin tribromide), and water or a lower alcohol (col. 1, lines 1-4; col. 6, 57-71; col. 7, lines 1-6; and col. 7, lines 21-26). The Examiner states that because Furusaki et al. teach the benefit of adding PVA to help improve film formation in an ITO film, it is obvious to one of ordinary skill to modify the composition of Olson to incorporate the polyvinyl alcohol of Furusaki et al.

However, Furusaki et al. teach that because in the previous research, the method in which an ITO thin film is prepared by using organometallic compounds of indium and tin as starting materials resulted in a high specific resistivity caused by carbon remaining in the film due to the large amount of organic substances contained in the coating liquid, Furusaki et al. then used a sol of colloidal particles formed from inorganic salts for the purpose of preparing ITO films having excellent electroconductivities while protecting the films from the influence of organic substances (page 1, line 24 to page 2, line 1 of the translation as attached). Although Furusaki et al. teach the use of polyvinyl alcohol as an auxiliary agent (page 3, lines 8-10 of the translation as attached), the teaching of Furusaki et al. is directed to adding polyvinyl alcohol in the preparation of the ITO films from the starting material of inorganic salts, in order to avoid the influence caused by the organic substances (page 1, line 19 to page 2, line 6 of the translation as attached). Furusaki et al. clearly teach to use inorganic salts instead of organometallic compounds of indium and tin as starting materials. Thus, the teaching of Furusaki et al. should be a teaching away from combining organometallic compounds of indium and tin as starting materials as taught by Olson.

(ii) Please compare Example 1 with Comparative Example 1 at pages 51 and 54 of the specification. This comparison shows that the surface resistivity is decreased to about 1/3 in the present invention by using a water-soluble organic high molecular weight compound. This significant decrease should be considered unexpected results. Furusaki et al. do not disclose or

Amendment  
Application No. 10/602,906  
Attorney Docket No. 052455

suggest that the surface resistivity of the films can be decreased by using a water-soluble organic high molecular weight compound when a transparent electroconductive film is formed with an organometallic compound starting material. Thus, the invention of claim 1 is not obvious over Olson in view of Furusaki.

(iii) To the dependent claims 2 to 7, 10, and 11, the same arguments as explained above are applied. In addition, claim 5, as amended, recites that a water-soluble organic high molecular weight compound is contained at a content of 1 to 10 wt% based on the total contents of the solution for preparing a transparent electroconductive film. Due to the feature, the surface resistivity of the film as prepared can be effectively decreased. Furusaki et al. do not disclose or suggest the range of the water-soluble organic high molecular weight compound content of the present invention, for the purpose of decreasing the surface resistivity. Thus, even a combination of the cited references does not make the invention of claim 5.

(2) Claims 8 and 9 were rejected under 35 U.S.C. §103(a) as being unpatentable over Olson and Furusaki et al., and further in view of Yudasaka et al. (U.S. Patent Application No. 2002/0074547 A1).

Claims 8 and 9 depend on claim 1. None of the cited reference teaches the motivation to combine the references. Reconsideration of the rejection is respectfully requested.

Amendment  
Application No. 10/602,906  
Attorney Docket No. 052455

(3) As to claims 12-16, none of the cited references teaches the motivation to combine the references to obtain the claimed invention.

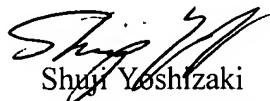
(4) In view of the aforementioned amendments and accompanying remarks, Applicant submits that the claims, as herein amended, are in condition for allowance. Applicant requests such action at an early date.

If the Examiner believes that this application is not now in condition for allowance, the Examiner is requested to contact Applicant's undersigned representative at the telephone number indicated below to arrange for an interview to expedite the disposition of this case.

If this paper is not timely filed, Applicant respectfully petitions for an appropriate extension of time. The fees for such an extension or any other fees that may be due with respect to this paper may be charged to Deposit Account No. 50-2866.

Respectfully submitted,

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Attachment: Limited Recognition  
Amendment Transmittal  
Petition for Extension of Time  
Partial Translation of Furusaki et al.



**PARTIAL TRANSLATION OF**

***Journal of the Ceramic Society of Japan 102 [2] 200-205(1994)***

***Preparation of ITO Thin Films by Sol-Gel Method***

5 ***Tsuyoshi FURUSAKI, Junichi TAKAHASHI and Kohei KODAIRA***

*Page 200, left column, line 19 to page 201, left column, line 5*

10 1. Introduction

Indium oxide thin films containing a small amount of tin (ITO: Indium-Tin-Oxide) have high electrical conductivity and are transparent to visible lights. Due to such properties, ITO films are widely applied to transparent electrodes of liquid crystal displays and plasma displays such as those typically found in watches, calculators and the like. Thus, ITO films are one of the essential materials in the field of optoelectronics.

ITO thin films are generally prepared by vacuum evaporation<sup>1)2)</sup>, sputtering methods<sup>3)4)</sup> and the like. In addition to these methods, a dip-coating method can be mentioned, which can easily form ITO thin films at low cost on large area substrates, such as transparent electrodes of solar batteries, heat ray reflecting films, etc. The authors have prepared an ITO thin film using organometallic compounds of indium and tin as a starting material by a coating method and reported the properties<sup>5)</sup> thereof. Since this method use a coating liquid containing a large amount of organic substance(s), carbon remains in the prepared film. Thus, the specific resistivity of the films is an order of magnitude higher ( $6 \text{ to } 8 \times 10^{-3} \Omega \cdot \text{cm}$ ) than that of thin films prepared by other methods. The authors attempted to use a sol of colloidal particles formed from inorganic salt to prepare ITO thin films for the purpose of preparing ITO films having excellent electrical conductivity while protecting the films from the influence of organic

substances. When an ITO film is prepared from a sol of colloidal particles formed by adding ammonia water to an aqueous solution of indium sulfate and tin sulfate, it was found that the ITO film prepared exhibited a specific resistivity of  $2$  to  $3 \times 10^{-3}$   $\Omega \cdot \text{cm}$  and 90% or higher transmittance over the visible light range<sup>6)</sup>.

In the case that colloidal particles are prepared from inorganic salt, the particle diameter and shape of colloidal particles noticeably vary depending on the type of salt as a starting material. Due to the rarity of reports on the preparation of thin films using colloidal particles, the relationship between the properties of colloidal particles and the properties of thin films prepared therefrom is not clear; however, the properties of colloidal particles and thin films prepared therefrom are possibly closely related. This study aims to prepare an ITO thin film by a sol-gel method using a sol of colloidal particles formed from indium nitrate as indium salt, and thus the following examinations are conducted:

- (1) examining the properties of colloidal particles prepared from indium nitrate;
- (2) investigating the effects of indium chloride essential for dispersion of colloidal particles, and examining the production process of an ITO thin film at the early stage of firing; and
- (3) examining the properties of ITO thin films prepared from colloidal particles formed from indium nitrate.

## 2. Experimental Procedure

Commercially available indium nitrate ( $\text{In}(\text{NO}_3)_3 \cdot 3\text{H}_2\text{O}$ ) and anhydrous tin chloride ( $\text{SnCl}_4$ ) were used as starting materials. Tin chloride was added to a 0.14 M aqueous indium nitrate solution to a concentration of 14 mol% relative to the concentration of Indium nitrate. Then, 2M ammonia water was used to titrate the solution to a pH of 8.5, so that the colloidal particles of indium hydroxide (hereafter referred to as "mother particles") were precipitated. After the mother particles

were washed by decantation, centrifugation was carried out (6500 rpm, 20 min.), and a sol was prepared by ultrasonic dispersion in a 0.28 M aqueous indium chloride solution or in a 0.84 M aqueous hydrochloric acid solution. Meanwhile, the volume of these aqueous solutions for dispersing the mother particles was set to 1/10 the volume of the 0.14 M aqueous indium nitrate solution. After adding 2.7 M acetic acid to the sol as a gelation inhibitor, polyvinyl alcohol was added as an auxiliary agent for film formation (PVA; added to 0.3 wt% of the coating solution), and thus the coating solution was prepared. The concentration of the mother particles in the coating solution was about 16 wt% calculated as  $\text{In}_2\text{O}_3$ .

A glass slide or a silica glass substrate was immersed in the coating solution, pulled up at the speed of 1 to 27 cm/min, dried at 55°C and 110°C each for 30 minutes, to form a gel film, and then fired at 550°C for 2 to 30 minutes to obtain an ITO thin film. A gel film formed from a sol with no auxiliary agent for film formation separated from the substrate or showed cracks. Thus, PVA<sup>6) to 8)</sup>, whose effect as an auxiliary agent for film formation was confirmed, was added to the sol, thereby preventing formation of cracks, etc.

*Page 204, right column, line 15 to page 205, left column, line 4*

The above results showed that, after firing for about 2 minutes, thermal decomposition of the mother particles and indium chloride is complete, developing a significant degree of a thin film microstructure. However, a longer firing duration may be required for preparation of ITO thin films suitable for practical use, considering the fact that the electrical conductivity is remarkably influenced by their crystallinity, solid solution of  $\text{Sn}^{4+}$  into the  $\text{In}^{3+}$  site, etc. Fig. 10 shows the effect of firing duration at 550°C on the specific resistivities of ITO thin films. With two-minute firing in which thermal decomposition is essentially complete,



so that a thin film microstructure is developed, the specific resistivity of the thin film is extremely high. As the firing duration increases, the specific resistivity decreases, and after firing for 20 minutes or longer, the resistivity became constant. Thus, it was found that firing for 20 minutes or longer was necessary for the preparation of an ITO thin film having excellent electrical conductivity. The specific resistivity of a thin film prepared by firing at 550°C for 30 minutes was  $5 \times 10^{-2} \Omega \cdot \text{cm}$ , which was approximately two orders of magnitude higher than that of thin films prepared by vacuum evaporation<sup>11)</sup>. This may be because the ITO thin films prepared in this study are porous. Considering the fact that ITO thin films with thickness up to 2  $\mu\text{m}$  can be prepared in a single dipping-firing procedure and the sheet resistivity of the thin film is about 500  $\Omega/\text{cm}^2$ , such a thin film can be put into practical use depending on the intended use. When such a thin film is heated at 300°C for 30 minutes *in vacuo* (about  $10^{-3}$  Torr), the sheet resistivity further decreases to reach 150 to 200  $\Omega/\text{cm}^2$ . The light transmittance for visible light of the thin film was 90% or higher.